IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:
Jin Kinoshita, Moritsugu Morita, Minehiro Mori and Yoichi Kodama
Group Art Unit: 1712
Application No: 10/512,064
Examiner: Kuo-Liang Peng
Filed: (PCT) May 29, 2003:
For: ADHESIVE RESIN AND FILM ADHESIVE MADE BY USING THE SAME

DECLARATION
PURSUANT TO 37 C.F.R. 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Kazunori Kamio, do declare and state as follows:

I graduated from Kyoto University, with a Bachelor's degree in Industrial Chemistry in March 1999, and a Master's Degree in Fundamental Energy Science in March 2001;

I joined Mitsui Chemicals, Inc. in April 2001;

From October 2001 to July 2003, I was engaged in research in the field of computational science at the Computational Science Research Room at the Material Science Laboratory;

From July 2003 to April 2005, I was engaged in research in the field of polymer simulation at the National Technical University of Athens, Greece; and

Since April 2005, I have been engaged in research in the field of computational science at the Computational Science Research Room at the Material Science Laboratory.

I am familiar with the Office Action dated August 22, 2006, and understand the Examiner's rejection therein.

The following additional simulations were carried out by me, on behalf of the inventors, in order to make the advantages of the subject matter clearer, on December 26, 2006, December 27, 2006 and February 7, 2007, at the Computational Science Research Room at the Material Science Laboratory, Mitsui Chemicals, Inc., Sodegaura Branch, at 580-32, Nagaura, Sodegaura-shi, Chiba, 299-0265, JAPAN.

Purpose of Simulation

In order to determine the effectiveness of use of the diamine structure of amended claim 8 of the present invention in the formation of a polyimide having a lower glass transition temperature (Tg), a computational simulation was conducted to compare the Tg of the sample polyimides [P1] to [P5].

The respective sample polyimides were obtained by polymerizing an acid anhydride ODPA (4,4'-oxydiphtalic anhydride) with a diamine having a structure of amended claim 8 of the present invention ([P1]) and other diamine structures disclosed in the cited references ([P2] to [P5]).

The polyimides [P2] and [P3] have a diamine structure disclosed in JP-A No. 2000-143981. [P2] has the amino group in the meta position in common with [P1] in consideration of JP '981 teaching that "the amino group is preferably in the meta position", but the other linkage groups are all in the para position. [P3] has all the linkage groups including the amino group in the para position.

The Tg of the polyimide having a diamine structure disclosed in JP-A No. 08-134213 ([P4]) and the polyimide having a diamine structure disclosed in JP-A No. 08-127656 ([P5]) were also simulated for comparison.

Simulation Method

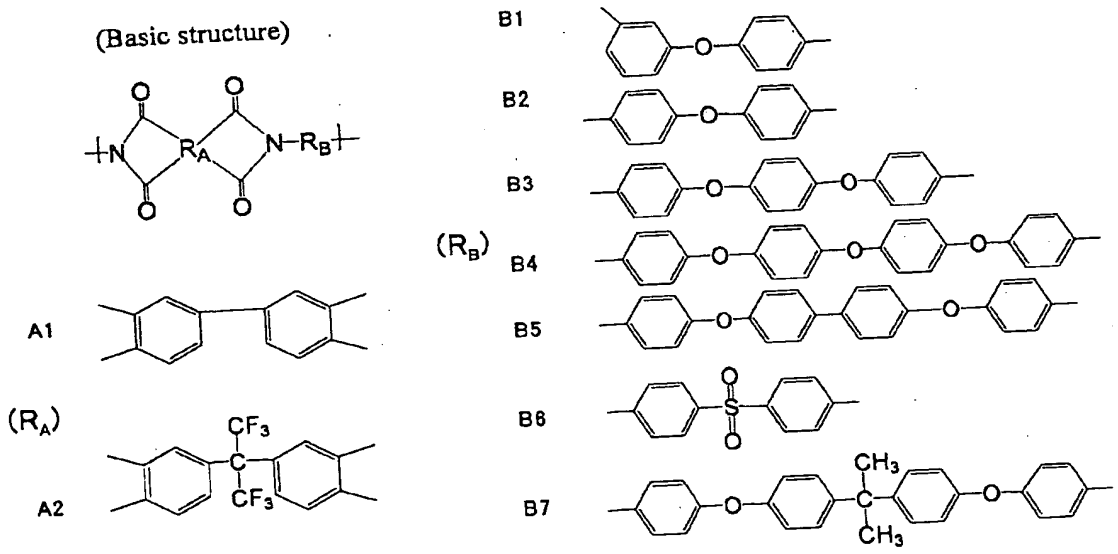
The glass transition temperature (Tg) of the sample polyimides was calculated using the Synthia Module [1] included in the program that can calculate the polymer properties in accordance with a quantitative structure-property relationships (Materials Studio v4.0.0, developed by Accelrys Software Inc.). The

Module is developed based on a method described in Bicerano [2] and is capable of calculating various properties such as Tg from the molecular structure of the polymers.

Preliminary Simulation for Determination of the Method

The Tgs of six BPDA polyimides and two 6FDA polyimides were simulated using the Synthia Module and compared with the experimentally obtained Tgs thereof that have been disclosed in the documents [3] and [4]. The chemical structures that constitute the polyimides 1 to 8 used for the preliminary simulation are shown in Fig. 1.

Fig. 1



In Fig. 1, R_A represents an acid anhydride-derived structure (A1: BPDA; A2: 6FDA) and R_B represents a diamine-derived structure (B1:3,4'-ODA; B2:4,4'-ODA; B3:1,4-4APB; B4:4,4'-4APDE; B5:4,4'-4APBPh; B6: 4,4'-DDS; B7:2,2-BAPP).

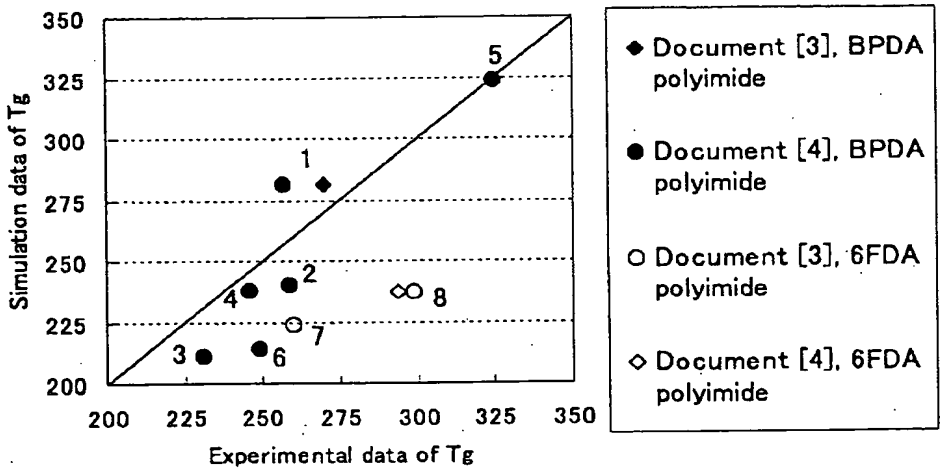
The combination of the acid anhydride and diamine, experimental data of the Tg in the documents [3] and [4], and the simulation data of the Tg of the respective polyimides 1 to 8 are shown in Table 1.

Table 1

Polyimide			Glass transition temperature		
	Acid anhydride	Diamine	Document [3]	Document [4]	Simulation
1	(A1) BPDA	(B2) 4,4'-ODA	270	257	281
2	(A1) BPDA	(B3) 1,4-4APB	-	259	240
3	(A1) BPDA	(B4) 4,4'-4APDE	-	231	211
4	(A1) BPDA	(B5) 4,4'-4APBPh	-	246	238
5	(A1) BPDA	(B6) 4,4'-DDS		325	324
6	(A1) BPDA	(B7) 2,2'-BAPP		249	214
7	(A2) 6FDA	(B1) 3,4'-ODA	260	-	224
8	(A2) 6FDA	(B2) 4,4'-ODA	299	294	237

The correlation between the experimental data of the Tg in documents [3] and [4] and the simulation data of the Tg are shown in Fig. 2. The numbers in Fig. 2 correspond to the sequential numbers attached to the polyimides as above.

Fig. 2



The results of the above preliminary simulation can be summarized as follows.

1. The simulation data of the four BPDA polyimides consisting of the acid anhydride structure (A1) and the respective diamine structures (B2) to (B5), comprising only benzene and ether oxygen, conformed to the experimental data with an accuracy in the range of about 20°C.

2. A trend was observed in which the Tg of the polyimide decreases as the number of the benzene ring-ether oxygen linkage increases, from the simulation data of the three BPDA polyimides with a combination of the structures (A1)-(B2), (A1)-(B3) and (A1)-(B4), which is consistent with the trend in the experimental data.

3. A trend was observed in which the Tg of the polyimide decreases as the number of the ether oxygen increases, from the simulation data of the BPDA polyimides with a combination of the structures (A1)-(B4) and (A1)-(B5), which is consistent with the trend in the experimental data.

4. A result was obtained in which the polyimide having a linkage in the meta position has a lower Tg than the polyimide having the linkage in the para position, from the simulation data of the 6FDA polyimides having the structure of (A2)-(B1) and (A2)-(B2), which is consistent with the result in the experimental data.

5. The simulation data of the polyimide (A1)-(B6) having a $-SO_2-$ linkage showed a high degree of consistency with the experimental data.

6. The simulation data of the 6FDA polyimides having the structure of (A2) and the polyimide having the structure of (B7) showed a rather low degree of consistency with the experimental data.

In view of the above results, it is suggested that the simulation data of the Tg of the polyimide that qualitatively conformed to the experimental data can be obtained by the method used in the above simulation.

Simulation of Tg of Polyimides [P1] to [P5]

Using the same method as above, the Tgs of five ODPA polyimides [P1] to [P5] having the chemical structure shown in Fig. 3 were calculated, wherein:

the polyimide [P1] has the diamine structure of amended claim 8 where the amino group is in the meta position and the linking groups between the benzene rings are all in the meta position;

the polyimide [P2] has an exemplary diamine structure of formula 2 in JP '981 where the amino group is in the meta position and the linking groups between the benzene rings are all in the para position;

the polyimide [P3] has an exemplary diamine structure of formula 2 in JP '981 where the amino group is in the para position and the linking groups between

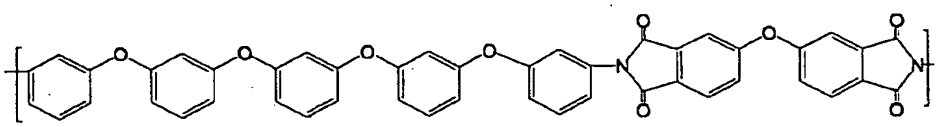
the benzene rings are all in the para position;

the polyimide [P4] has the diamine structure used in Example 1 of JP '213 (4,4'-bis(3-aminophenoxy)diphenyl ether) where the amino group is in the meta position and the linking groups between the benzene rings are all in the para position; and

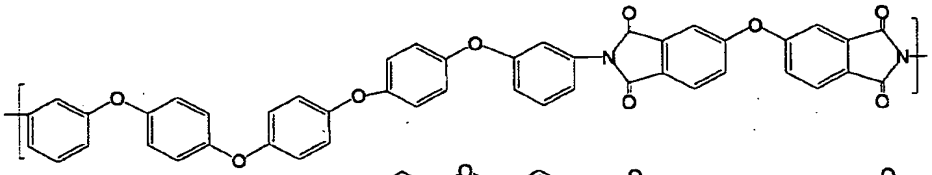
the polyimide [P5] has the diamine structure used in Example 4 of JP '656 (3,3'-bis[4-(3-aminocumyl)phenoxy]diphenyl sulfone) where the amino group is in the meta position and the linking groups between the benzene rings are partly in the meta position.

Fig. 3

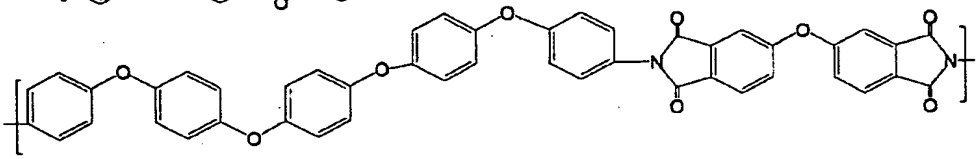
[P1]



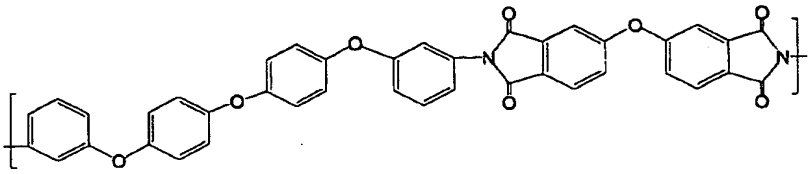
[P2]



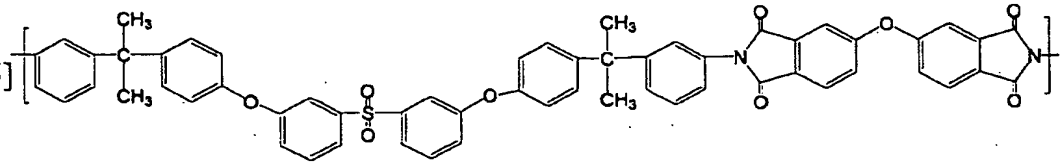
[P3]



[P4]



[P5]



The simulated Tgs of the above polyimides were 142°C ([P1]), 165°C ([P2]), 180°C ([P3]), 183°C ([P4]), and 208°C ([P5]), respectively. A trend was observed in which a polyimide having a diamine structure with more linkage groups in the meta position has a lower Tg.

The simulated Tg of [P5] was higher than that of the ODPA polyimides [P1] to [P4], which suggests that the experimentally obtained Tg of [P5] would be even higher than 208°C, in consideration of the result of the preliminary simulation (the polyimide having the diamine structure (B7) showed a lower simulated Tg than the experimentally obtained Tg).

Conclusion

The above results show that the diamine structure of amended claim 8 of the present invention is effective for the formation of a polyimide having a lower Tg.

Cited References

[1]http://www.accelrys.com/products/mstudio/modeling/polymersandsimulations/synthia.html

[2] Bicerano, J. Prediction of Polymer Properties, Third Edition, Marcel Dekker Inc.: New York, 2002.

[3] Tanaka, K.; Kita, H.; Okano, M.; Okamoto, K. Polymer 1992, 33, 585-592.

[4] Hirayama, Y.; Yoshinaga, T.; Kusuki, Y.; Ninomiya, K.; Sakakibara, T.; Tamari, T. J. Membr. Sci 1996, 111, 169-182.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

DATE: February 15, 2007

Kazunori Kamio

Kazunori KAMIO